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THEORY OF ABSORPTION LINE SHAPES IN MONATOMIC GASES;

I. GENERAL FORMULATION AND APPROXIMATE SOLUTIONS. *

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A formalism is developed for calculating the complex refractive index of a monatomic gas as a function of frequency in the region of an absorption. The treatment involves a direct study of the properties of the dressed photon state in the gas, and includes effects due to translational motion of the atoms as well as the dipolar "resonance" interactions between them. It is necessary to assume that the electromagnetic properties of the gas are describable in some detail by a linear frequency - and wave number-dependent susceptibility. A set of coupled nonlinear integral equations are derived which together determine the susceptibility function and hence the observable refractive index. The "static" limit of large atomic mass is considered in some detail, and a first correction to it is also obtained. The results are compared with measurements by Tomiser on the linewidths of the sodium D-line at various temperatures and pressures. We obtain qualitative agreement, while previous theoretical linewidths were too small by a factor of order 10^3.

I. INTRODUCTION

The problem of calculating absorption line shapes in monatomic gases, especially the contribution of the dipolar "resonance" interactions between different absorbers, has been attacked in the past in several different ways, all of a more or less ad hoc nature.[1] The various methods ("collision", "statistical") are in agreement among themselves at least as to the qualitative features of the line shape, and as to the order of magnitude of the linewidth. However, it appears to have gone largely unnoticed that they are in strong disagreement with the latest experimental results.[2,3] In particular, the calculated "resonance" width[1] is too small by a factor of the order of 10^3 to fit the experimental data, and the Doppler width is also much too small to account for the observed effects. (Moser and Schultz[3] give a detailed comparison of the theoretical results with their own and other experiments.)

One of us[4,5] has developed a theory of the line shape which begins from the more fundamental point of view of actually studying the properties of the stationary states of the quantum mechanical system composed of radiation field and matter. This theory, however, considered each absorber to occupy a fixed position in space, so that effects due to the motion of the atoms were left out. The original formulation depended on the use of

[1] R. G. Breene, Jr., Revs. Modern Phys. 29, 94 (1957)

[2] J. Tomiser, Acta Phys. Austriaca 8, 198 (1953); 8, 276 (1954); 8, 323 (1954); 9, 18 (1954).

3 H. Moser and H. Schultz, Ann. Physik 7, 243 (1959).

4 C. A. Mead, Phys. Rev. 120, 854 (1960).

5 C. A. Mead, Phys. Rev. 120, 860 (1960).

"damping operators"[6], but more recently the results have been rederived by means of a simpler formalism, and at the same time brought into rough agreement with experiment.[7]

The purpose of the present article is to remove the defect of neglecting the translational motion of the absorbers, and also to discuss in somewhat more detail than before the comparison of theory with experiment. Our method will be a generalization of that of reference 7; that is, we will study the properties of the stationary states of the system but will also assume that the system is describable by means of a refractive index. This assumption will be generalized to take account of the possibility of "spatial dispersion".[8] The plan of the article is as follows: In section II, the notation is introduced and some preliminary matters discussed, including the "cutoff function" inserted in certain Hamiltonian matrix elements which is supposed to represent roughly the effect of finite atomic size and which later prevents certain integrals from diverging. In section III, the basic assumption that the medium is describable in terms of a refractive index is stated precisely as a statement about the stationary states of the system. Section IV consists of the derivation of a set of integral equations which determine the refractive index. Section V discusses the "static limit" of large atomic mass (which corresponds to the situation in references 4, 5, and 7) and also the lowest order correction for finite mass. It is shown that under the conditions of Tomiser's experiments on Sodium[2] the static approximation should be adequate. Section VI gives a more detailed comparison with Tomiser's results in the static limit,

6 C. A. Mead, Phys. Rev. 112, 1843 (1958).

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including the effect of the spin-orbit splitting. We give a detailed
comparison only with Tomiser's experiments on the Sodium D-line; the
situation with the other experiments of references 2 and 3 is qualitatively
similar. A brief discussion of the cutoff function used is given in an
appendix.

A subsequent paper by one of us will discuss another method for
obtaining values for certain integral properties of the line shape, including
corrections for the failure of one of the assumptions of Sec. III.

II. NOTATION AND HAMILTONIAN

Our system consits of N identical absorbers, each with mechanical
mass m, distributed in a volume V, within which the radiation field is
required formally to obey periodic boundary conditions. In the "initial"
state $\left| 0 \right>$, each atom A has a momentum $\hbar q_A$ assigned to it, and the
number of atoms with momentum in a given region of wave-number space
is described by a Boltzmann distribution at some temperature T. For the
sake of definiteness, the ground electronic state of each atom is considered
to be a nondegenerate s-state, and the only excited level (or at least the
only one of interest to us) is assumed to be a triply degenerate p-level.
The energy difference between the two levels is $\hbar \nu_o$. A given state of
the system of radiation field plus matter may differ from the state $\left| 0 \right>$ in
one or more of three ways: (1) There may be a photon present; this is
denoted by a greek letter, e. g. λ denotes the presence of a bare photon of
wave number κ_λ and polarization ϵ_λ. (2) One or more of the atoms may
be excited; this is indicated by a capital Latin letter denoting which atom is
excited, with a subscript giving the direction of "polarization" (electric
dipole moment matrix element between excited state and ground state).
Thus A_i represents the excitation of atom A, with dipole moment matrix

element in the i-direction (i = x, y, z). (3) One or more of the atoms may have acquired a momentum different from that assigned to it in the state $|0\rangle$. $\underset{\sim}{\kappa}$ (A) indicates that atom A has an excess momentum $\hbar\underset{\sim}{\kappa}$, so that its total momentum is $\hbar(\underset{\sim}{q}_A + \underset{\sim}{\kappa})$. The way in which these symbols are combined in specifying a state is best explained by means of a few examples of states represented in our notation and with their properties also written out in words:

$\left|\eta, \underset{\sim}{\kappa}(A)\right\rangle$: Photon present with wave number $\underset{\sim}{\kappa}_\eta$ and polarization $\underset{\sim}{\epsilon}_\eta$; atom A has excess momentum $\underset{\sim}{\kappa}$; all other atoms as in state $|0\rangle$.

$\left|Ai, \underset{\sim}{\kappa}_1(A), \underset{\sim}{\kappa}_2(B)\right\rangle$: Atom A excited, with "polarization" in i-direction; atom A has excess momentum $\hbar\underset{\sim}{\kappa}_1$; atom B has excess of $\hbar \underset{\sim}{\kappa}_2$; all other atoms as in state $|0\rangle$; no photons present.

We are interested in constructing the "dressed" photon state $|\bar{\lambda}\rangle$, which is an eigenstate of the complete Hamiltonian with energy $\hbar\nu_\lambda$ ($\nu_\lambda = c\kappa_\lambda$), and which in some sense approaches the bare photon state $|\lambda\rangle$ in the limit of zero interaction. We assume that $|\bar{\lambda}\rangle$ can be built up entirely out of two kinds of states:

(1) States with one photon present, and no electronic excitation.

(2) States with one and only one atom excited, and no photons present.

States coming under categories (1) and (2) may still, of course, include any combination of excess translational momenta for the various atoms. Other states, such as two-photon states and states with several atoms excited, may be included later by means of perturbation theory, if desired.

The approximation of limiting ourselves to the states of types (1) and (2) should hold in the frequency region $\nu_\lambda \cong \nu_o$, which is the region we are interested in here. It could be made more formally precise by means of the Arnous-Bleuler transformation,[4,9,10] but this would make some of the subsequent manipulations more complicated so it was decided not to use it in this article.

9 E. Arnous and K. Bleuler, Helv. Phys. Acta 25, 581 (1952).

10 W. Heitler, The Quantum Theory of Radiation (Oxford University Press, New York, 1954), 3'rd ed, pages 348-353.

The refractive index can be thought of as describing the propagation of the electric field matrix element

$$\langle 0|E_o(R)|\bar{\lambda}\rangle = \mathcal{E}_\gamma \langle 0|E_o(R)|\gamma\rangle\langle\gamma|\bar{\lambda}\rangle .$$

This immediately leads to a difficulty: It would appear that because of momentum conservation $\langle \gamma|\bar{\lambda}\rangle = 0$ unless $\hbar_\gamma = \hbar_\lambda$. This would mean that

$$\langle 0|E_o(R)|\bar{\lambda}\rangle = E_{oo} \exp(i\, \hbar_\lambda \cdot R),$$

corresponding to a refractive index of unity. This problem does not arise in the situation treated in references 4, 5, and 7, since there the momentum conservation is spoiled by holding the atoms fixed. This apparently hopeless difficulty really is the result of a rather trivial effect: Momentum is indeed conserved when an electromagnetic wave propagates through a medium with altered wave number, but the excess momentum is taken up either by the medium as a whole or by the walls containing the medium. Since explicit inclusion of the effects of the walls in a calculation is not

feasible, we will treat the excess momentum as belonging to the medium as a whole. From this point of view, the propagation of the field should be considered relative to the center of mass of the medium. Therefore, instead of

$$\underset{\sim}{E}_O(\underset{\sim}{R}) = \sum_{\gamma} \underset{\sim}{\epsilon}_{\gamma} \left\{ s_{\gamma} \exp(i \underset{\sim}{\mathcal{K}}_{\gamma} \cdot \underset{\sim}{R}) + s_{\gamma}^{+} \exp(-i \underset{\sim}{\mathcal{K}}_{\gamma} \cdot \underset{\sim}{R}) \right\}$$

(where s_{γ} and s_{γ}^{+} are respectively destruction and creation operators, apart from constants), we use

$$\underset{\sim}{E}(r) = \underset{\sim}{E}_O(\frac{1}{N} \sum_{A} \underset{\sim}{r}_A + \underset{\sim}{r})$$

$$= \sum_{\gamma} \underset{\sim}{\epsilon}_{\gamma} \left\{ s_{\gamma} \exp(i \underset{\sim}{\mathcal{K}}_{\gamma} \cdot \underset{\sim}{r}) \prod_A \exp(i \frac{\underset{\sim}{\mathcal{K}}}{N}_{\gamma} \cdot \underset{\sim}{r}_A) + s_{\gamma}^{+} \exp(-i \underset{\sim}{\mathcal{K}}_{\gamma} \underset{\sim}{r}) \prod_A \exp(-i \frac{\underset{\sim}{\mathcal{K}}}{N}_{\gamma} \cdot \underset{\sim}{r}_A) \right\}$$

When $\underset{\sim}{E}(\underset{\sim}{R})$ is applied to a state, it not only creates (destroys) a photon, but also alters the momenta of all the atoms by a small and equal amount in such a way that total momentum is conserved. From now on, therefore, when we write down a state it is to be understood that, in addition to the photons, excitations, etc. specifically noted, the state also includes an additional superimposed momentum of the entire medium (equally divided between the N atoms) such that the total momentum of the state is zero. With this understanding all the states we are talking about have the same momentum, and we can study the propagation of

$$\langle 0 | \underset{\sim}{E}(\underset{\sim}{r}) | \bar{\lambda} \rangle = \sum_{\gamma} \langle 0 | \underset{\sim}{E}(\underset{\sim}{r}) | \gamma \rangle \langle \gamma | \bar{\lambda} \rangle .$$

There will be no need to mention this explicitly again. Indeed, most of the subsequent manipulations could be gone through formally without performing this trick at all, the only trouble being that they would be meaningless due to both sides of the various equations being zero. The energy associated with this superimposed motion is, of course, negligible in the limit $N \rightarrow \infty$.

We now give some typical Hamiltonian matrix elements. All the ones we will need will differ from these at most by the presence of an

excess momentum on an atom appearing on both sides of the matrix element and having no effect except to shift the energy in the case of a diagonal matrix element. The energy of the state $|\,0\rangle$ is taken as zero. We obtain the following matrix elements for the Hamiltonian operator $\mathcal{H}$:

$$\langle Ai, \underset{\sim}{\varkappa}(A)|\mathcal{H}|Ai, \underset{\sim}{\varkappa}(A)\rangle = \hbar\,[\nu_0 + K(\underset{\sim A}{q}, \underset{\sim}{\varkappa})]\ , \tag{1}$$

where

$$K(\underset{\sim}{q}, \underset{\sim}{\varkappa}) = \frac{\hbar}{2m}\,[2\,\underset{\sim}{q}\cdot\underset{\sim}{\varkappa} + \varkappa^2]\ ; \tag{2}$$

$$\langle \lambda|\mathcal{H}|\eta\rangle = \hbar\,\nu_\lambda\,\delta_{\lambda\eta}\ ; \tag{3}$$

$$\langle Ai, \underset{\sim}{\varkappa}\eta\,(A)|\mathcal{H}|\eta\rangle = -\frac{2\,i\,\pi^{1/2}}{V^{1/2}}\,\nu_0\,\mu\,\epsilon_\eta\,i\left(\frac{\hbar}{2\nu_\eta}\right)^{1/2}\,G^{1/2}(\varkappa_\eta)\,; \tag{4}$$

$$\langle Ai, \underset{\sim}{\varkappa}_1(A)|\mathcal{H}|Bj, \underset{\sim}{\varkappa}(B), (\underset{\sim}{\varkappa}_1 - \underset{\sim}{\varkappa})(A)\rangle = \frac{4\pi}{V}\,\mu^2\,G(\varkappa)\,\frac{\varkappa_i\,\varkappa_j}{\varkappa^2} \tag{5}$$

In equations (4) and (5), μ is the absolute value of the dipole moment matrix element for the transition, and $G(\varkappa)$ is a cutoff factor which we now pause to discuss.

Our state $|\,0\rangle$ has each atom in a definite momentum state, and we are not including any interactions in which two atoms can exchange momentum with both remaining in the ground state; that is, we are neglecting direct interactions between atoms in the ground electronic state. If these interactions were included, their main effect would be to introduce a correlation in the positions of the atoms such that the distance between the centers of mass of two atoms can never be less than an atomic diameter. We take this into account by altering the interaction so that, while atoms are formally permitted to approach each other arbitrarily closely, they are not permitted to exchange electronic excitation except at distances

greater than the atomic diameter r_0. In this way, the transfer of excitation should proceed just as if there were no pairs of atoms closer to one another than r_0. In momentum space, this means that the momentum transfer accompanying the exchange of electronic excitation should not be greater in order of magnitude than $\hbar(1/r_0)$, and this is embodied in the factor $G(\kappa)$. Thus, G is a function which is nearly unity when $\kappa \ll 1/r_0$, and becomes very small or zero when $\kappa \gg 1/r_0$. For the Gaussian cutoff factor used later on in this article, a way of relating the momentum space cutoff to a particular value of r_0 is given in the appendix. This prescription is not unique, however.

We will use the remainder of this section to dispose of one more preliminary matter. Later on we will have occasion to evaluate the quantity

$$W = \lim_{\xi \to 0+} \sum_{\eta} \frac{|\langle Ai, \underset{\sim}{\kappa}(A)| \mathcal{H} | \eta, (\underset{\sim}{\kappa} - \underset{\sim}{\kappa}\eta)(A)\rangle|^2}{\hbar[\nu_\lambda - \nu\eta - K(q_A, \underset{\sim}{n} - \underset{\sim}{\kappa}\eta) \pm i\xi]} \qquad (6)$$

In situations of interest to us here,

$$|\nu_\lambda - \nu_0| \ll \nu_0$$

Thus, we can estimate W by replacing ν_λ by ν_0. With this replacement, however, Re W is just the second-order transverse self-energy of the state $|Ai, \underset{\sim}{\kappa}(A)\rangle$, and may therefore be absorbed into ν_0 by renormalization. If we also assume

$$K(\underset{\sim}{q} A, \underset{\sim}{\kappa} - \underset{\sim}{\kappa}\eta) \ll \nu_0$$

and remember that $G(\nu_0/c) \cong 1$, then we get (replacing the sum over η by integration)

$$W = -i\hbar\sigma = -\frac{2}{3} i \frac{\nu_0^3}{c^3} \mu^2, \qquad (7)$$

where σ is the natural linewidth. In what follows, we will always use (7).
The approximations involved are: (1) Neglect of recoil corrections to the
natural linewidth, which are small and can be included if desired; (2)
Neglect of of the dependence of Im W on ν_λ, which is also very small in
the frequency region of interest; (3) Neglect of the cutoff-dependent Re W
when $\nu_\lambda \neq \nu_o$. The formal justification of this requires the Arnous-Bleuler
transformation.[4,9,10] If the reader will accept this one result, he will be
spared the complications that this transformation would introduce in the
subsequent manipulations.

III. BASIC ASSUMPTIONS

Classically, one may expand the electric field $\underset{\sim}{E}$ and Polarization
$\underset{\sim}{P}$ in Fourier series as follows:

$$\underset{\sim}{E}(r,t) = \int_{-\infty}^{\infty} d\nu \sum_{\alpha} E_\alpha(\nu)\, \underset{\sim}{\epsilon}_\alpha\, \exp i[\underset{\sim}{k}_\alpha \cdot \underset{\sim}{r} - \nu t] \; ;$$

$$\underset{\sim}{P}(r,t) = \int_{-\infty}^{\infty} d\nu \sum_{\alpha} P_\alpha(\nu)\, \underset{\sim}{\epsilon}_\alpha\, \exp i[\underset{\sim}{k}_\alpha \cdot \underset{\sim}{r} - \nu t].$$

Here the notation α represents a wave number $\underset{\sim}{k}_\alpha$ and a polarization unit
vector $\underset{\sim}{\epsilon}_\alpha$; for each wave number there are three polarizations, two
transverse and one longitudinal. The allowed wave numbers are those
compatible with the periodic boundary conditions.

The requirements that the medium be isotropic and describable by
a refractive index (but with the possibility of "spatial dispersion") may be
expressed by

$$4\pi\, P_\alpha(\nu) = F(\nu, k_\alpha, \underset{\sim}{k}_\alpha \cdot \underset{\sim}{\epsilon}_\alpha)\, E_\alpha(\nu), \tag{8}$$

where F is a function depending only on the frequency ν, the absolute value
k of the wave number, and whether the mode α is longitudinal or trans-
verse. We will also use the notations $F_t(\nu, \kappa)$, $F_\ell(\nu, \kappa)$ to indicate the trans-

verse and longitudinal susceptibilities respectively.

Quantum mechanically, $\underset{\sim}{E}$ and $\underset{\sim}{P}$ become operators, but otherwise the situation is unchanged. In the Schrödinger Picture, the frequencies are to be replaced by energy differences divided by $\hbar$, and we have different equations for the various matrix elements of the operators. A quantum analog of eq. (8) is

$$\langle 0 \mid 4\pi P_{\alpha} \mid \bar{\lambda} \rangle = F(\nu_{\lambda}, \, \eta_{\alpha}, \eta_{\alpha} \cdot \xi_{\alpha}) \, \langle 0 \mid E_{\alpha} \mid \bar{\lambda} \rangle. \qquad (9)$$

It will be convenient to rearrange (9) slightly before using it. We note that

$$\underset{\sim}{E} = \underset{\sim}{E}_{coul} - \frac{1}{c} \, \frac{\partial}{\partial t} \underset{\sim}{A}$$

where $\underset{\sim}{E}_{coul}$ is the electrostatic longitudinal Coulomb field, and $\underset{\sim}{A}$ is the transverse vector potential (we use Coulomb Gauge throughout). We also have

$$\langle 0 \mid -\frac{1}{c} \, \frac{\partial}{\partial t} \underset{\sim}{A} \mid \bar{\lambda} \rangle = \frac{i\nu_{\lambda}}{c} \, \langle 0 \mid \underset{\sim}{A} \mid \bar{\lambda} \rangle. \qquad (10)$$

Hence if we define

$$\underset{\sim}{\mathcal{E}} = \underset{\sim}{E}_{coul} + \frac{i\nu_{\lambda}}{c} \, \underset{\sim}{A},$$

then we can combine (9), (10), and (11) to obtain

$$\langle 0 \mid 4\pi P_{\alpha} \mid \bar{\lambda} \rangle = F(\nu_{\lambda}, \, \eta_{\alpha}, \, \eta_{\alpha} \cdot \mathcal{E}_{\alpha}) \langle 0 \mid \mathcal{E}_{\alpha} \mid \bar{\lambda} \rangle. \qquad (12)$$

Eq. (12) is the expression we will use for the requirement that the medium be describable by a refractive index.

It is also useful to notice that the electrostatic dipole-dipole interaction, whose matrix element is given by (5) may be written as

$$\mathcal{H}_{AB} = -\underset{\sim}{\mu}_A \cdot \underset{\sim}{E}_{coul} \, (B, A),$$

where $\underset{\sim}{\mu}_A$ is the electric dipole moment operator of atom A and $\underset{\sim}{E}_{coul}$ (B, A) is the electrostatic field produced by atom B at the position of atom A. We can therefore write

$$\langle Ai, \underset{\sim}{\mathcal{K}}(A)| \mathcal{H}|Bj, \underset{\sim}{\mathcal{K}}_{\mathcal{L}}(B), (\underset{\sim}{\mathcal{K}} - \underset{\sim}{\mathcal{K}}_{\mathcal{L}})(A)\rangle$$
$$= -\mathcal{H} \frac{\mathcal{K}_{\mathcal{L}i}}{\mathcal{K}_{\mathcal{L}}}\langle 0| \mathcal{E}_{\mathcal{L}}|Bj, \underset{\sim}{\mathcal{K}}_{\mathcal{L}}(B)\rangle. \tag{13}$$

We also note that eq. (4) can be rearranged as follows:

$$\langle Aj, \underset{\sim}{\mathcal{K}} \gamma (A)| \mathcal{H}|\gamma\rangle = -i \frac{\nu_0}{c} \mu \epsilon \gamma j \langle 0| \Delta \gamma|\gamma\rangle$$
$$= -\mu \epsilon \gamma j \frac{\nu_0}{\nu_\lambda} \langle 0| \mathcal{E}_\gamma|\gamma\rangle$$
$$\cong -\mu \epsilon \gamma j \langle 0| \mathcal{E}_\gamma|\gamma\rangle, \tag{14}$$

since we are interested only in the situation where

$$|\nu_\lambda - \nu_0| << \nu_0. \tag{15}$$

In order to obtain a closed set of integral equations to determine the functions F, we will also need a stronger assumption which is a generalization of (12):

$$\langle \underset{\sim}{K}(A)| 4\pi P_*^{(A)}|\bar{\lambda}\rangle = F[\nu_\lambda - K(\underset{\sim}{q}_A; \underset{\sim}{K}), \mathcal{K}_{\mathcal{L}} \cdot \underset{\sim}{\mathcal{K}}_{\mathcal{L}} \cdot \mathcal{E}_{\mathcal{L}}]\langle \underset{\sim}{\mathcal{K}}(A)| \mathcal{E}_{\mathcal{L}}|\bar{\lambda}\rangle. \tag{16}$$

where $\underset{\sim}{P}^{(A)}$ is the polarization of the medium due to all atoms except atom A. To understand the meaning of (16), suppose that the initial state of atom A is not known with certainty, so that the "initial" state of the medium is

not simply $|0\rangle$, but

$$|0'\rangle = |0\rangle + u\,|\underset{\sim}{\cancel{k}} (A)\rangle .$$

Now the matrix element $\langle 0'|\underset{\sim}{\xi_k}|\bar\lambda\rangle$ will have a term of frequency $\nu_\lambda -$ $K(\underset{\sim}{q_A}, \underset{\sim}{\cancel{k}})$ equal to $u^*\langle\underset{\sim}{\eta}(A\underset{\sim}{\cancel{k}}|\bar\lambda\rangle$, and a similar situation will hold for $\underset{\sim}{P}^{(A)}$. Equation (16) expresses the assumption that the polarization of the rest of the medium is still related to the field (which now includes contributions from the virtual excitations of atom A) by the same susceptibility function. Eq. (16) does represent an approximation beyond (12); Classically, we can always limit our considerations to a linear susceptibility simply by making our fields arbitrarily small; but in quantum mechanics we cannot make the matrix elements appearing in (12) become small. Their values are fixed by the solution of the eigenvalue problem. Nevertheless, we can define a function F by means of (12), and this will lead to no contradictions as long as we restrict ouselves to those matrix elements in (12). Moreover, this is the obvious quantum mechanical analog to the classical procedure of allowing the fields to approach zero: the one-photon state is as close to zero as we can get and still have any field at all. Strict linearity would be expressed by requiring that expressions such as (12) hold for all matrix elements of the operators involved, and in particular for those appearing in (16). Hence, (16) is a stronger linearity assumption than (12). In this article we will assume that it holds, but in the next article it will be shown how the failure of (16) to hold exactly can lead to changes in the value of the function F appearing in (12).

IV. INTEGRAL EQUATIONS FOR F

In order to find expressions for the various matrix elements appearing in eqs. (12), (16), we must know something about the dressed photon state $|\bar{\lambda}\rangle$. Denoting general states by a, b, etc., we have

$$\hbar (\nu_\lambda - \nu_a) \langle a | \bar{\lambda} \rangle = \mathcal{E}_{b \neq a} \langle a | \mathcal{H} | b \rangle \langle b | \bar{\lambda} \rangle, \tag{17}$$

where $\hbar \nu_a$ is the energy of the zero-order state $|a\rangle$. The appropriate "retarded" solution of (17) is

$$\langle a | \bar{\lambda} \rangle = \hbar^{-1} (\nu_\lambda - \nu_a + i\zeta)^{-1} \mathcal{E}_{b \neq a} \langle a | \mathcal{H} | b \rangle \langle b | \bar{\lambda} \rangle \tag{18}$$

in which the limit $\zeta \longrightarrow 0^+$ is always understood.

Now let us examine some field and polarization matrix elements. If η is a transverse mode, so that there are photons of type η , we have

$$\langle 0 | \mathcal{E}_\eta | \bar{\lambda} \rangle = \langle 0 | \mathcal{E}_\eta | \eta \rangle \langle \eta | \bar{\lambda} \rangle \tag{19}$$

For the polarization we have

$$\underset{\sim}{P}(r) = \sum_A \mu_{\sim A} \int (\underset{\sim}{r} - \underset{\sim}{r}_A),$$

$$P_\eta = \frac{1}{V} \int \underset{\sim}{\epsilon}_\eta \cdot \underset{\sim}{P}(r) \exp(-i \underset{\sim}{K}_\eta \cdot \underset{\sim}{r}) d^3 r$$

$$= \frac{1}{V} \sum_A \underset{\sim}{\epsilon}_\eta \cdot \mu_{\sim A} \exp(-i \underset{\sim}{K}_\eta \cdot \underset{\sim}{r}_A),$$

so that

$$\langle 0 | 4\pi P_\gamma | \bar{\lambda} \rangle = \mathcal{E}_A \langle 0 | 4\pi P_\gamma | Aj, \underset{\sim}{k}\gamma (A) \rangle \langle Aj, \underset{\sim}{k}\gamma (A) | \bar{\lambda} \rangle$$

$$= \frac{4\pi\mu}{V} \mathcal{E}_A \epsilon \gamma j \langle Aj, \underset{\sim}{k}\gamma (A) | \bar{\lambda} \rangle . \qquad (20)$$

Here and henceforth a summation over repeated coordinate indices j, k, etc. is understood. We will now apply (17) to $| a \rangle = | Aj, \underset{\sim}{k}\gamma (A) \rangle$, and also use (14). The result is

$$\hbar [\omega - K (\underset{\sim A}{q}, \underset{\sim}{k}\gamma)] \langle Aj, \underset{\sim}{k}\gamma (A) | \bar{\lambda} \rangle = -\mu \epsilon \gamma j \langle 0 | \mathcal{E}_\gamma | \gamma \rangle \langle \gamma | \bar{\lambda} \rangle$$

$$+ \mathcal{E}_{s \neq \gamma} \langle Aj, \underset{\sim}{k}\gamma (A) | \mathcal{H} | S, (\underset{\sim}{k}\gamma - \underset{\sim}{k}s)(A) \rangle \langle S, (\underset{\sim}{k}\gamma - \underset{\sim}{k}s)(A) | \bar{\lambda} \rangle \qquad (20a)$$

$$+ \mathcal{E}^{(\ell)}_{B \neq A, \angle} \langle Aj, \underset{\sim}{k}\gamma (A) | \mathcal{H} | B_\ell, (\underset{\sim}{k}\gamma - \underset{\sim}{k}_\angle)(A), \underset{\sim}{k}_\angle (B) \rangle \langle B_\ell, (\underset{\sim}{k}\gamma - \underset{\sim}{k}_\angle)(A) \underset{\sim}{k}_\angle (B) | \bar{\lambda} \rangle$$

Here the last summation goes over all longitudinal modes $\angle$, and we have defined

$$\omega \equiv \nu_\lambda - \nu_0$$

We now apply (18) to the state $| a \rangle = | S, (\underset{\sim}{k}\gamma - \underset{\sim}{k}s)(A) \rangle$, and also use eqs. (6), (7), and (19). The result is

$$(21)$$

$$\hbar [w - K (\underset{\sim A}{q}, \underset{\sim}{k}\gamma)] \langle Aj, \underset{\sim}{k}\gamma (A) | \bar{\lambda} \rangle = -\mu \xi \gamma j \langle 0 | \mathcal{E}_\gamma | \bar{\lambda} \rangle - i \sigma \langle Aj, \underset{\sim}{k}\gamma (A) | \bar{\lambda} \rangle$$

$$+ \mathcal{E}_{B \neq A, \angle} \langle Aj, \underset{\sim}{k}\gamma (A) | \mathcal{K} | B_\ell, (\underset{\sim}{k}\gamma - \underset{\sim}{k}_\angle)(A), \underset{\sim}{k}_\angle (B) \rangle \langle B_\ell, (\underset{\sim}{k}\gamma - \underset{\sim}{k}_\angle)(A), \underset{\sim}{k}_\angle (B) | \bar{\lambda} \rangle.$$

The last summation now goes over all modes, transverse and longitudinal. In arriving at (21), we have assumed that the omission of the one photon mode γ from the summation in (6) does not affect the result. The matrix

elements of $\mathcal{H}$ are defined by

$$\langle Aj, \underset{\sim}{\mathcal{H}}(A)|K|B_{\ell}, \underset{\sim\ell}{\mathcal{H}}(B), (\underset{\sim}{\mathcal{H}}-\underset{\sim\ell}{\mathcal{H}})(A)\rangle = \langle Aj, \underset{\sim}{\mathcal{H}}(A)|\mathcal{H}|B_{\ell}, \underset{\sim\ell}{\mathcal{H}}(B), (\underset{\sim}{\mathcal{H}}-\underset{\sim\ell}{\mathcal{H}})(A)\rangle$$

$$+ \frac{\langle Aj, \underset{\sim}{K}(A)|\mathcal{H}|\mathcal{L}, (\underset{\sim}{\mathcal{H}}-\underset{\sim\ell}{\mathcal{H}})(A)\rangle \langle \mathcal{L}, \underset{\sim}{\mathcal{H}}-\underset{\sim\ell}{\mathcal{H}}(A)|\mathcal{H}|B_{\ell}, \underset{\sim\ell}{\mathcal{H}}(B), (\underset{\sim}{\mathcal{H}}-\underset{\sim\ell}{\mathcal{H}})(A)\rangle}{\hbar [\nu_{\lambda}-\nu_{\mathcal{L}} - K(\underset{\sim A}{q}, \underset{\sim}{\mathcal{H}}-\underset{\sim\ell}{\mathcal{H}})) + i \zeta]} \tag{22}$$

The first term on the right-hand side of (22) is nonzero only for longitudinal modes, the second only for transverse modes.

We now make the following definition:

$$-i\hbar\sigma\langle Aj, \underset{\sim}{\mathcal{H}}\gamma(A)|\bar{\lambda}\rangle + \underset{B\neq A, \mathcal{L}}{\Sigma}\langle Aj, \underset{\sim}{\mathcal{H}}\gamma(A)|K|B_{\ell}, (\underset{\sim}{\mathcal{H}}\gamma-\underset{\sim\ell}{\mathcal{H}})(A), \underset{\sim\ell}{\mathcal{H}}(B)\rangle \times$$
$$\times \langle Bk, (\underset{\sim}{\mathcal{H}}\gamma-\underset{\sim\ell}{\mathcal{H}})(A), \underset{\sim\ell}{\mathcal{H}}(B)|\bar{\lambda}\rangle$$

$$\equiv \hbar \Delta jk(\omega, \underset{\sim}{\mathcal{H}}\gamma, \underset{\sim A}{q})\langle Ak, \underset{\sim}{\mathcal{H}}\gamma(A)|\bar{\lambda}\rangle . \tag{23}$$

Now eq. (23) can be inserted into (21), and the result solved for $\langle Aj, \underset{\sim}{\mathcal{H}}\gamma(A)|\bar{\lambda}\rangle$. The result is

$$\langle Aj, \underset{\sim}{\mathcal{H}}\gamma(A)|\bar{\lambda}\rangle = -\frac{\mu}{\hbar}\langle 0|\mathcal{E}_{\mathcal{H}}|\bar{\lambda}\rangle [\omega - K(\underset{\sim A}{q}, \underset{\sim}{\mathcal{H}}\gamma)-\Delta]_{jk}^{-1}\mathcal{E}_{\mathcal{H}}k \tag{24}$$

The matrix $U = (\omega - K - \Delta)^{-1}$ is, of course, the inverse of the matrix whose elements are

$$(\omega - K)\delta_j k - \Delta jk.$$

It is now a simple matter to substitute (24) into (20), sum over A using the Boltzmann distribution, and solve eq. (12) for F_t with the aid of the

result. This gives the solution

(25)

$$F_t(\omega, \varkappa_\gamma) = - \frac{4\pi \mu^2}{\mu} \frac{N}{V} (\frac{\beta}{\pi})^{3/2} \int \exp(-\beta q^2) \varepsilon_{\gamma j} [\omega - K(q, \varkappa_\gamma) - \Delta(\omega, \varkappa_\gamma, q)]^{-1}_{jk\chi} \times \varepsilon_{\gamma k} \ell^3_{j}$$

where

$$\beta = \frac{\mu^2}{2\ mkT} \quad , \quad \text{and k is Boltzmann's constant.}$$

We could have replaced the matrix product under the integral sign by

$$\varepsilon_{\gamma j}\ U_{jk}\ \varepsilon_{\gamma k} = \frac{1}{2}[U_{kk} - \frac{1}{\varkappa^2}\ \varkappa_j\ U_{jk}\ \varkappa_k].$$

It will also be necessary to derive an expression analogous to (25)
for the longitudinal field. If $\mathcal{L}$ is a longitude mode,

$$\langle 0 | \mathcal{E}_{\mathcal{L}} | \bar{\lambda} \rangle = \mathcal{E}_A \langle 0 | \mathcal{E}_{\mathcal{L}} | Aj, \varkappa_{\mathcal{L}} (A) \times Aj, \varkappa_{\mathcal{L}} (A) | \bar{\lambda} \rangle, \quad (26)$$

and

$$\langle 0 | 4\pi P_{\mathcal{L}} | \bar{\lambda} \rangle = \frac{4\pi \mu}{V} \mathcal{E}_B \frac{\varkappa_{\mathcal{L}} k}{\varkappa_{\mathcal{L}}} \langle \beta k, \varkappa_{\mathcal{L}} (B) | \bar{\lambda} \rangle \quad (27)$$

Now eq s. (17), (18) may be used much as before to give

$$k \cdot [\omega - K(q_B, \varkappa_{\mathcal{L}})] \langle \beta k, \varkappa_{\mathcal{L}} (B) | \bar{\lambda} \rangle = \mathcal{E}_{A \neq B} \langle \beta k, \varkappa_{\mathcal{L}} (B) | \mathcal{H} | A_{\ell} \varkappa_{\mathcal{L}} (A) \times A_{\ell} \varkappa_{\mathcal{L}} (A) | \lambda \rangle$$

$$- i k \sigma \langle \beta k, \varkappa_{\mathcal{L}} (B) | \bar{\lambda} \rangle.$$

$$+ \mathcal{E}_{A \neq B, \gamma \neq \mathcal{L}} \langle \beta k, \varkappa_{\mathcal{L}} (B) | \mathcal{H} | A_{\ell} \varkappa_{\gamma} (A), (\varkappa_{\mathcal{L}} - \varkappa_{\gamma})(B) \rangle \langle A_{\ell} \varkappa_{\gamma} (A), (\varkappa_{\mathcal{L}} - \varkappa_{\gamma})(B) | \bar{\lambda} \rangle.$$

By using (6), (7), (13), (22), (23) in a manner similar to the derivation of (21), we easily find

$$\mathcal{X} \cdot [w - K(q_B, \mathcal{K}_C)] \langle \beta k, \mathcal{K}_C (B) | \bar{\lambda} \rangle = -\mu \frac{\mathcal{K}_C \, k}{\mathcal{K}_C} \langle 0 | \mathcal{E}_\mathcal{L} | \bar{\lambda} \rangle$$
$$+ \mathcal{X} \, \Delta_{kl}(\omega, \mathcal{K}_C , q_B) \langle \beta_l, \mathcal{K}_C (B) | \bar{\lambda} \rangle , \tag{28}$$

in which we have also made the assumption that the omission of one atom from the sum in (26) will not noticeably affect the result. Now one proceeds just as in the derivation of (25): Solve (28) for $\langle B_k, \mathcal{K}_C (B) | \bar{\lambda} \rangle$, insert the result in (27), carry out the summation (replacing it by integration over the Boltzmann distribution), and solve eq. (12) for F_ℓ. The result is

$$F_\ell(\omega, \mathcal{K}) = - \frac{4\pi\mu^2}{\mathcal{X}} \frac{N}{V} \left(\frac{\beta}{\pi}\right)^{3/2} \int \exp(-\beta q^2) \frac{Kj}{\mathcal{K}} [\omega - K(q, \mathcal{K}) - \Delta(\omega, \mathcal{K}_A q)]_{jk}^{-1}$$
$$\frac{\mathcal{K} k}{\mathcal{K}} d^3 q. \tag{29}$$

Eqs. (25) and (29) determine the two functions F if Δ_{jk} can be determined. We must now turn our attention to the evaluation of Δ. There are (as far as we know) two methods for doing this. One uses a formal infinite series expansion and is perfectly general if the series converges. The other gives the result in closed form, but requires the assumption (16) which has not been used up to now.

(1) Formal Expansion

By comparing eqs. (20a), (21), (23) it is easy to see that

$$\mathcal{X} \, \Delta_{jk}(\omega, \mathcal{K}\gamma , q_A) \langle A_k, \mathcal{K}\gamma (A) | \bar{\lambda} \rangle$$
$$= \sum_a \langle Ak, \mathcal{K}\gamma (A) | \mathcal{H} | a \rangle \langle a | \bar{\lambda} \rangle . \tag{30}$$

The summation goes over all states except $| a \rangle = | \gamma \rangle$. Since there is no singular contribution from $| \gamma \rangle$, it may be considered to go over all states.

The next step is to substitute (18) into the right-hand side of (30). Then use (18) again to replace all the terms $\langle b | \bar{\lambda} \rangle$ on the right-hand

side of the result, except for $|b\rangle = |A_k, \underset{\sim}{\kappa}(A)\rangle$, and repeat this indefinitely. This gives

$$\hbar\, \Delta_{jk}(\omega, \underset{\sim}{\kappa}, \underset{\sim}{q})\langle A_k, \underset{\sim}{\kappa}(A)|\bar{\lambda}\rangle$$

$$\sum_a' \sum_b \frac{\langle Aj, \underset{\sim}{\kappa}(A)|\mathcal{H}|a\rangle\langle a|\mathcal{H}|b\rangle\langle b|\bar{\lambda}\rangle}{\hbar(\nu_\lambda - \nu_a + i\xi)}$$

$$= \sum_a' \frac{\langle Aj, \underset{\sim}{\kappa}(A)|\mathcal{H}|a\rangle\langle a|\mathcal{H}|A_k, \underset{\sim}{\kappa}(A)\rangle\langle A_k, \underset{\sim}{\kappa}(a)|\bar{\lambda}\rangle}{\hbar(\nu_\lambda - \nu_a + i\xi)}$$

$$+ \sum_a' \sum_b' \frac{\langle Aj, \underset{\sim}{\kappa}(A)|\mathcal{H}|a\rangle\langle a|\mathcal{H}|b\rangle\langle b|\bar{\lambda}\rangle}{\hbar(\nu_\lambda - \nu_a + i\xi)}$$

$$= \sum_a' \frac{\langle Aj, \underset{\sim}{\kappa}(A)|\mathcal{H}|a\rangle\langle a|\mathcal{H}|A_k, \underset{\sim}{\kappa}(A)\rangle\langle A_k, \underset{\sim}{\kappa}(A)|\bar{\lambda}\rangle}{\hbar(\nu_\lambda - \nu_a + i\xi)}$$

$$+ \sum_{a,b}' \frac{\langle Aj, \underset{\sim}{\kappa}(A)|\mathcal{H}|a\rangle\langle a|\mathcal{H}|b\rangle\langle b|\mathcal{H}|A_k, \underset{\sim}{\kappa}(A)\rangle\langle A_k, \underset{\sim}{\kappa}(A)|\bar{\lambda}\rangle}{\hbar^2(\nu_\lambda - \nu_a + i\xi)(\nu_\lambda - \nu_b + i\xi)} + --(31)$$

where a prime on a summation means that the three states $|A_k, \underset{\sim}{\kappa}(A)\rangle$ are to be ommitted ($k = x, y, z$). By inspection of this series, we see that Δ_{jk} can be expressed formally as

$$\hbar\, \Delta_{jk}(\omega, \underset{\sim}{\kappa}, \underset{\sim}{q}_A) = \sum_a' \frac{\langle Aj, \underset{\sim}{\kappa}(A)|\mathcal{H}|a\rangle\langle a|\mathcal{H}|A_k, \underset{\sim}{\kappa}(A)\rangle}{\hbar(\nu_\lambda - \nu_a + i\xi)}$$

$$+ \sum_{a,b}' \frac{\langle Aj, \underset{\sim}{\kappa}(A)|\mathcal{H}|a\rangle\langle a|\mathcal{H}|b\rangle\langle b|\mathcal{H}|A_k, \underset{\sim}{\kappa}(A)\rangle}{\hbar^2(\nu_\lambda - \nu_a + i\xi)(\nu_\lambda - \nu_b + i\xi)} + ---- \qquad (32)$$

The expansion (32) will be made use of in the following paper. For the remainder of this article, however, we will make use of a formal closed form solution which depends on the assumption (16).

(2) Closed Integral Equation Using (16)

We first consider the contribution of the longitudinal coulomb part of $\mathcal{K}$ to Δ_{jk}. Denoting this by a superscript ℓ, and using eqs. (5), (22), and (23), we can express this term as

$$\mathbf{1}\,\Delta_{jk}^{\ell}(w, \underset{M}{\mathcal{K}}, q_A) \langle A_k, \underset{M}{\mathcal{K}}(A) | \bar{\lambda} \rangle \equiv$$

$$\equiv \sum_{B \neq A, \mathcal{L}} \langle Aj, \underset{M}{\mathcal{K}}(A) | \mathcal{H} | \beta k, \underset{M\mathcal{L}}{\mathcal{K}}(B), (\underset{M}{\mathcal{K}} - \underset{M\mathcal{L}}{\mathcal{K}})(A) \rangle \langle \beta k, \underset{M\mathcal{L}}{\mathcal{K}}(B), (\underset{M}{\mathcal{K}} - \underset{M\mathcal{L}}{\mathcal{K}})(A) | \bar{\lambda} \rangle$$

$$= \frac{4\pi}{V} \mu^2 \sum_{\mathcal{L}} G(\mathcal{K}_{\mathcal{L}}) \frac{\mathcal{K}_{\mathcal{L}j}}{\mathcal{K}_{\mathcal{L}}} \int_{\mathcal{L}}^{\ell(A)} \tag{33}$$

where

$$\int_{\mathcal{L}}^{\ell(A)} = \sum_{B \neq A} \frac{\mathcal{K}_{\mathcal{L}k}}{\mathcal{K}_{\mathcal{L}}} \langle \beta k, \underset{M\mathcal{L}}{\mathcal{K}}(B), (\underset{M}{\mathcal{K}} - \underset{M\mathcal{L}}{\mathcal{K}})(A) | \bar{\lambda} \rangle,$$

and the sum over $\mathcal{L}$, of course, includes only longitudinal modes. Using (5), (13), and (33), we can now obtain

$$\langle (\underset{M}{\mathcal{K}} - \underset{M\mathcal{L}}{\mathcal{K}})(A) | \mathcal{E}_{\mathcal{L}} | \bar{\lambda} \rangle = \langle (\underset{M}{\mathcal{K}} - \underset{M\mathcal{L}}{\mathcal{K}})(A) | \mathcal{E}_{\mathcal{L}} | A_k, \underset{M}{\mathcal{K}}(A) \rangle \langle A_k, \underset{M}{\mathcal{K}}(A) | \bar{\lambda} \rangle$$

$$+ \sum_{B \neq A} \langle (\underset{M}{\mathcal{K}} - \underset{M\mathcal{L}}{\mathcal{K}})(A) | \mathcal{E}_{\mathcal{L}} | \beta k, \underset{M\mathcal{L}}{\mathcal{K}}(B), (\underset{M}{\mathcal{K}} - \underset{M\mathcal{L}}{\mathcal{K}})(A) \rangle \; \times$$

$$\times \langle B_k, \underset{M\mathcal{L}}{\mathcal{K}}(B), (\underset{M}{\mathcal{K}} - \underset{M\mathcal{L}}{\mathcal{K}})(A) | \bar{\lambda} \rangle$$

$$= -\frac{4\pi}{V} \mu G(\mathcal{K}_{\mathcal{L}}) \frac{\mathcal{K}_{\mathcal{L}k}}{\mathcal{K}_{\mathcal{L}}} \langle A_k, \underset{M\mathcal{L}}{\mathcal{K}}(A) | \bar{\lambda} \rangle - \frac{4\pi}{V} \mu G(\mathcal{K}_{\mathcal{L}}) \int_{\mathcal{L}}^{\ell(A)}$$

$$\tag{34}$$

It is also evident that

$$\left\langle (\nu \underset{\sim}{\ell} - \underset{\sim}{\ell}\mathcal{L})(A) \,\middle|\, 4\pi P_{\mathcal{L}}^{(A)} \,\middle|\, \bar{\lambda} \right\rangle = \frac{4\pi\mu}{V} \int_{\mathcal{L}}^{\ell\,(A)} \tag{35}$$

It is now a simple matter to insert (34) and (35) into (16) and solve for $\int_{\alpha}^{\ell(A)}$:

$$\int_{\mathcal{L}}^{\ell\,(A)} = - \frac{G(\kappa_{\mathcal{L}}) F_{\ell}[\omega - K(q_A, \kappa - \kappa_{\mathcal{L}}), \kappa_{\mathcal{L}}] \frac{\kappa_{\mathcal{L}} k}{\kappa_{\mathcal{L}}} \left\langle A_k, \kappa(A) \,\middle|\, \bar{\lambda} \right\rangle}{1 + G(\kappa_{\mathcal{L}}) F_{\ell}[\omega - K(q_A, \kappa - \kappa_{\mathcal{L}}), \kappa_{\mathcal{L}}]} \tag{36}$$

The next step is to substitute (36) into (33), replace the sum over $\mathcal{L}$ by integration, and extract the value of Δ_{jk}. The result is

$$\not{k} \Delta_{jk}^{\ell}(w, \kappa, q) = - \frac{\mu^2}{2\pi^2} \int \frac{G^2(\kappa') F_{\ell}[\omega - K(q, \kappa - \kappa'), \kappa'] \frac{\kappa'_j \kappa'_k}{\kappa'^2} d^3\kappa'}{1 + G(\kappa') F_{\ell}[\omega - K(q, \kappa - \kappa'), \kappa']} \tag{37}$$

There is one more contribution to Δ_{jk}, namely that due to the transverse part of $\mathcal{K}$. It can be written as

$$\not{k} \Delta_{jk}^{t}(\omega, \kappa, q_A) \left\langle A_k, \kappa(A) \,\middle|\, \bar{\lambda} \right\rangle$$

$$\equiv \sum_{B \neq A, \gamma} \frac{2\pi}{V} \frac{v_0^2 \mu^2 G(\kappa\gamma) \epsilon_{\gamma j} \epsilon_{\gamma k} \left\langle Bk, (\kappa - \kappa_\gamma)(A), \kappa_\gamma(B) \,\middle|\, \bar{\lambda} \right\rangle}{v_\gamma [v_\lambda - v_\gamma - K(q_A, \kappa - \kappa_\gamma) + i \mathfrak{z}]}$$

$$\equiv \frac{2\pi}{V} \sum_{\gamma} \frac{v_0^2 \mu^2 G(\kappa\gamma) \epsilon_{\gamma j}}{v_\gamma [v_\lambda - v_\gamma - K(q_A, \kappa - \kappa_\gamma) + i \mathfrak{z}]} \int_{\gamma}^{t\,(A)} \tag{38}$$

where

$$\int_{\gamma}^{t\,(A)} \equiv \sum_{B \neq A} \epsilon_{\gamma k} \left\langle Bk, (\kappa - \kappa_\gamma)(A), \kappa_\gamma(B) \,\middle|\, \bar{\lambda} \right\rangle$$

and we have used (22), (23), plus an obvious generalization of (4). We now proceed much as in the derivation of (37). First, by using (4), (14), and (18), we find

$$
\begin{aligned}
\langle (\underset{\sim}{k}-\underset{\sim}{\kappa}\gamma)(A) | \mathcal{E}\gamma | \bar{\lambda} \rangle &= \langle (\underset{\sim}{k}-\underset{\sim}{\kappa}\gamma)(A) | \mathcal{E}\gamma | \gamma, (\underset{\sim}{k}-\underset{\sim}{\kappa}\gamma)(A) \rangle \langle \gamma, \underset{\sim}{k}-\underset{\sim}{\kappa}\gamma)(A) | \bar{\lambda} \rangle \\
&= \langle 0 | \mathcal{E}\gamma | \gamma \rangle \langle \gamma, (\underset{\sim}{k}-\underset{\sim}{\kappa}\gamma)(A) | \bar{\lambda} \rangle \\
&= -\frac{\mu | \langle 0 | \mathcal{E}\gamma | \gamma \rangle |^2 [\in \gamma \, k \langle A_k, \underset{\sim}{\kappa}(A) | \bar{\lambda} \rangle + \Gamma_\gamma^{t\,(A)}]}{\mathbf{1} [v_\lambda - v\gamma - K(\underset{\sim}{q}_A, \underset{\sim}{\kappa}-\underset{\sim}{\kappa}\gamma) \pm i \mathcal{S}]} \quad (39)
\end{aligned}
$$

We also have, obviously,

$$
\langle (\underset{\sim}{\kappa}-\underset{\sim}{\kappa}\gamma)(A) | 4\pi P\gamma (A) | \bar{\lambda} \rangle = \frac{4\pi\mu}{V} \Gamma_\gamma^{t(A)} \quad (40)
$$

Now one substitutes (39) and (40) into (16), solves for $\Gamma_\gamma^{t\,(A)}$, inserts the result into (38), and again extracts the value of Δ^t. The result is

$$
\mathbf{1}\, \Delta_{jk}^t (\omega, \underset{\sim}{\kappa}, \underset{\sim}{q}) \quad (41)
$$

$$
= -\frac{v_o^4 \mu^2}{8\pi^2} \int \frac{[\int_{jk} - \frac{\kappa'_j \kappa'_k}{\kappa'^2}]\, G^2(\kappa')F_t[\omega - K(\underset{\sim}{q}, \underset{\sim}{\kappa}-\underset{\sim}{\kappa}'), \kappa'] d^3\kappa'}{\left(c^2\kappa'^2 (v_\lambda - c\kappa' - K(q, \kappa-\kappa') + i\mathcal{S})^2 \right.}
$$
$$
\left. + \frac{v_o^2\, G(\kappa') c\, \kappa' F_t[w - K(q, \kappa-\kappa'), \kappa']}{} \times \right.
$$
$$
\left. \times (v_\lambda - c\kappa' - K(q, \kappa \cdot \kappa' + i\mathcal{S}) \right)
$$

where we have also used (4) and (14) to get the matrix element of $\mathcal{E}$.

The matrix elements of Δ are now given by

$$\Delta_{jk} = -i \sigma \, \delta_{jk} + \Delta_{jk}^{\ell} + \Delta_{jk}^{t} \tag{42}$$

Equations (25), (29), (37), (41), and (42) now determine both functions F. The observed refractive index ρ is determined by the requirements that, first, the electromagnetic waves propagating in the absence of macroscopic charges are purely transverse; and second, that the usual relation between wave length and susceptibility holds. Thus we write

$$\rho^2 (\omega) - 1 = F_t(\omega, \rho^{\nu}\lambda/c) = F_t(\omega, \rho^{\nu}o/c). \tag{43}$$

The solution of (43) will in general require the analytic extension of F_t into the upper half of the complex k -plane.

V. STATIC LIMIT AND LOWEST ORDER CORRECTION

In the previous section, a set of coupled integral equations was derived which in principle determines the refractive index if the assumption (16) is valid. However, due to their highly nonlinear character (among other things), it is evident that a closed form solution of the equations is not feasible. Therefore, suitable approximations must be found. The reader will remember that we are particularly interested in the experimental conditions prevailing in Tomiser's experiments on sodium vapor. It will be recalled that the observed linewidth in these experiments was much greater than the Doppler width, suggesting that the effects of translational motion of the absorbers is not very important; this corresponds to the limit $m \rightarrow \infty$, which we call the "static limit." This limit is also of interest as a check on the work of references 4, 5, and 7, in which the absorbers were considered fixed in given positions in space.

In the static limit, all the quantities K become zero, and it is easy to see that Δ becomes a scalar:

$$\Delta_{jk} = \Delta_s \, \delta_{jk}$$

Because Δ is a scalar, we also have

$$F_t = F_\ell \equiv F_s,$$

and it further becomes possible to assume that F_s and Δ_s are both functions only of ω. We have from (25), (29), (37), (41), (42) in the static limit

$$F_s(\omega) = - \frac{4\pi\mu^2}{k} \frac{N}{V} \frac{1}{\omega - \Delta(\omega)} \tag{44}$$

and

$$\Delta_s(\omega) = - i\sigma + \Delta_s^\ell(\omega) + \Delta_s^t(\omega), \tag{45}$$

where

$$k \, \Delta_s^\ell(\omega) = - \frac{\mu^2}{2\pi^2} F_s(\omega) \frac{1}{3} \int \frac{G^2(k)d^3}{1 + G(k)F_s(w)} \tag{46}$$

and

$$k \, \Delta_s^t(\omega) = - \frac{v_o^4 \mu^2}{8\pi^2} \cdot \frac{2}{3} F_s(\omega) \int \frac{G^2(k)d^3 k}{c^2 k^2 (v_\lambda - ck + i\xi)^2 + \frac{v_o^2 G(k)}{2} F_s(\omega)k}$$

$$\times(ck) \, x(v_\lambda - ck + i\xi) \tag{47}$$

Equations (44) - (47) are essentially the same as the corresponding equations in references 5 and 7. There are two main differences: first, the use of the Arnous-Bleuler transformation brings about some small alterations in the denominator of (47), which will be discussed in more detail in the next section. Second, in the earlier work the cutoff function

G was left out until the very end and then inserted only when it was needed to prevent a divergence; hence, G does not appear in the analog of (47) at all, nor in the denominator of the analog of (46). Neither of these differences is very significant; the omission of the Arnous-Bleuler transformation and the way of bringing in the cutoff are both rather arbitrary, nonessential elements in the formalism. Since F_s is a function only of ω, we have in this limit $\rho^\lambda(\omega) - 1 = F_s(\omega)$.

We would now like to discuss the lowest order finite-mass correction to the static approximation, but this will be made easier if we first make another approximation. A comparison of (37) with (41) and (7) shows that

$$\Delta^l_{jk} >> \sigma \cdot \Delta^t_{jk}.$$

This is because only the integral (37) requires the cutoff for convergence, making Δ^l proportional to the third power of the cutoff wave number. For the remainder of this section, therefore, we will set Δ equal to Δ^l ; we will also omit the G in the denominator of (37) since, as pointed out above, the way in which the cutoff is brought in is rather arbitrary. Equations (29), (37) now become

(48)

$$F_l(\omega, \kappa) = - D(\beta/\pi)^{3/2} \int \exp(-\beta q^2) \frac{\kappa_j}{\kappa} [\omega - K(q, \kappa) - \Delta(\omega, \kappa, q]^{-1}_{jk} \frac{\kappa_k}{\kappa} d^3 q;$$

$$\Delta_{jk}(\omega, \kappa, q) = -B \int \frac{G^2(\kappa') F_l[\omega - K(q, \kappa - \kappa'), \kappa'] \frac{\kappa'_j \kappa'_k}{\kappa'^2} d^3 \kappa'}{1 + F_l[\omega - K(q, \kappa - \kappa'), \kappa']} \quad (49)$$

where

$$D = \frac{4\pi\mu^2}{\hbar} \frac{N}{V} ,$$

$$B = \frac{\mu^2}{2\pi^2 \hbar} , \quad (50)$$

We now expand F_ℓ and Δ_{jk} in inverse poweres of m, with the intention of evaluating the zero- and first-order terms. We write

$$F_\ell(\omega, \mathcal{K}) = F_s(\omega) + \frac{g(\omega, \mathcal{K})}{m} + \text{---} \tag{51}$$

and

$$\Delta_{jk}(\omega, \underset{\sim}{\mathcal{K}}, \underset{\sim}{q}) = \Delta s(\omega) \delta_{jk} + \frac{\gamma_{jk}(\omega, \underset{\sim}{\mathcal{K}}, \underset{\sim}{q})}{m} + \text{---} \tag{52}$$

It will now be convenient to use a specific form for the cutoff function. The choice made in this section is

$$G(\mathcal{K}) = \exp(-\mathcal{L}^2 \mathcal{K}^2)$$

According to the prescription given in the appendix, this corresponds to a cutoff radius of $r_0 = \mathcal{L} \pi^{1/2}$.

We now insert (51), (52) into (48) and (49), expand in powers of (1/m), and equate coefficients of like powers. By equating the zero-order coefficients we get

$$F_s(\omega) = - \frac{D}{\omega - \Delta_s(w)} \quad , \tag{53}$$

and

$$\Delta_s(\omega) = \frac{-\Lambda F_s(\omega)}{1 + F_s(\omega)} \quad , \tag{54}$$

where

$$\Lambda = \frac{B}{3} \left(\frac{\pi}{2\mathcal{L}^2}\right)^{3/2} \quad .$$

Equations (53), (54) can easily be solved, giving the result

$$F_s(\omega) = \frac{-(\omega + D) + [(w - D)^2 - 4D\Lambda]^{1/2}}{2(\omega + \Lambda)} \tag{55}$$

According to (55), F_s has an imaginary (absorptive) part in the region

$$|\omega - D| < 2(D \Lambda)^{1/2}$$

In this region, of course, the square root is taken so that the imaginary part is positive. In the regions to the left and right of the absorptive region, it is seen by analytic continuation, or by physical requirements, that one should take the negative square root on the left, the positive one on the right.

Since $\mathcal{L}$ is of the order of an atomic radius, in a dilute gas

$$\Lambda >> D .$$

Therefore, the denominator in (55) is a slowly varying function of ω in the absorptive region. If the denominator is considered approximately constant, we obtain for the half-width at half-height

$$\delta \omega_{1/2} = (3D\Lambda)^{1/2} = (\frac{\pi}{2})^{1/4} \frac{\mu^2}{\mu} (\frac{N}{V \mathcal{L}^3})^{1/2} . \tag{56}$$

Thus, the half-width should be proportional approximately to the square root of the density of the gas.

When we equate coefficients of the first power of $(1/m)$ in (49), we get

$$\gamma_{jk}(\omega, \mathcal{n}, q) = - \frac{\beta}{[1 + F_s(\omega)]^2} \int G^2(\mathcal{n}')d^3\mathcal{n}' \left\{ -F_s^{\ell}(\omega) \frac{\cancel{k}}{2}[2q \cdot (\mathcal{k} - \mathcal{k}') + (\mathcal{k} - \mathcal{k}')^2] + g(\omega, \mathcal{k}') \right\} \frac{\mathcal{k}'_j \mathcal{k}'_k}{\mathcal{n}'^2}$$

$$= - \frac{\beta \delta_{jk}}{3[1 + F_s(\omega)]^2} \left\{ -F_s^{\ell}(\omega) \frac{\cancel{k}}{2}(\frac{\pi}{2\mathcal{L}^2})^{3/2}[2q \cdot \mathcal{k} + \mathcal{k}^2 + \frac{3}{4\mathcal{L}^2}] + \zeta(\omega) \right\} \tag{57}$$

where

$$\mathcal{S}(\omega) = \int G^2(\mathcal{L}') \, g(\omega, \mathcal{L}') \, d^3 \mathcal{L}' \, . \tag{58}$$

In arriving at (57), we have made use of eq. (2) for K. The prime on F_s denotes differentiation with respect to ω. We can now insert (51) and (57) into (48), expand, and equate coefficients of (1/m):

$$g(\omega, \mathcal{L}) = a(\omega) + b(\omega) \mathcal{L}^2 + c(\omega) \, \mathcal{S}(\omega), \tag{59}$$

where

$$a(\omega) = - \frac{D \, B \, F_s'(\omega)}{[\omega - \Delta_s(\omega)] \, [1 + F_s(\omega)]^2} \, \frac{k}{8\mathcal{L}^2} \, (\frac{\pi}{2\mathcal{L}^2})^{3/2} \quad ;$$

$$b(w) = - \frac{D \, k}{2[\omega - \Delta_s(\omega)]^2} \left\{ 1 + \frac{B \, F_s'(\omega)}{3[1 + F_s(\omega)]^2} \, (\frac{\pi}{2\mathcal{L}^2})^{3/2} \right\} \tag{60}$$

$$c(\omega) = \frac{D \, B}{3[1 + F_s(\omega)]^2}$$

To evaluate $\mathcal{S}(\omega)$, we substitute (59) into (58), getting

$$\mathcal{S}(\omega) = \int G^2(\mathcal{L}') \, [a(\omega) + b(\omega) \mathcal{L}'^2 + c(\omega) \, \mathcal{S}(\omega)] \, d^3 \mathcal{L}'$$

$$= (\frac{\pi}{2\mathcal{L}^2})^{3/2} [a(\omega) + \frac{3 \, b(\omega)}{4\mathcal{L}^2} + c(\omega) \, \mathcal{S}(\omega)] \, . \tag{61}$$

Equation (61) is now solved for $\xi\ (\omega)$:

$$\xi(\omega) = \frac{(\frac{\pi}{2\alpha^2})^{3/2}[a(\omega) + \frac{3\ b(\omega)}{4\alpha^2}]}{1 - (\frac{\pi}{2\alpha^2})^{3/2}\ c(\omega)} \quad .$$

(62)

Equations (59), (60), and (62) determine g. Since Δ is still a scalar in this approximation, we still have $F_t = F_\ell$. Note that the result is still independent of T.

We have calculated $Im\ F_t(\omega,\ \nu_o/c)$ in both zero- and first-order under the conditions of one of Tomiser's experiments on sodium. F_t is sufficiently small in this case that we can neglect its dependence on wave number. The following parameters were used:

$$T = 688.1°\ K.$$
$$P = 0.503\ mm\ Hg.$$
$$\alpha = 2.83 \times 10^{-8}\ cm$$
$$\mu^2 = 4.167 \times 10^{-35}\ erg\ cm^3$$
$$\sigma = 3.199 \times 10^7\ radians/sec$$
$$\lambda = 2\pi/\nu_o = 5.89 \times 10^{-5}\ cm.$$

The results are shown in figure 1. Obviously, the lowest-order correction for finite mass has only a very small effect. If one looks closely enough at the figure, one can see that the finite-mass correction slightly raises the maximum of the line; it is therefore to be thought of as a narrowing effect. This can be understood qualitatively as follows: In the static approximation, the line is broadened due to the transfer of excitation from one absorber to another. This transfer may be accompanied by a momentum transfer of any size up to the cutoff momentum. When the finite mass is taken into account, however, excitation transfer accompanied by large momentum transfer is inhibited due to the recoil of the atoms spoiling the energy balance; i.e., excitation transfer with large momentum transfer is

no longer a "resonance" process. This effect might be called "recoil narrowing".

The half width at half maximum from fig. 1 is (in wave length units) 1.48 A. The experimental values are 1.565 A for the 3/2 line, and 1.075 for the 1/2. Thus, we are able to get qualitative agreement with experiment with a reasonable value for α. In the next section, we take into account explicitly the doublet character of the line, making possible a more detailed comparison with the experimental results.

VI. THE DOUBLET CASE IN STATIC APPROXIMATION

The sodium D-line is not actually a transition from a nondegenerate s-level to a triply degenerate p-level, as assumed up to now. The transition is actually from ^2S to ^2P, and the excited state has two components, $^2P_{1/2}$ and $^2P_{3/2}$. The wave lengths of the transitions are respectively 5896 Å and 5890 Å.

A detailed derivation of the formulas for the doublet case would be largely repititious. We will therefore content ourselves with rearranging some of the formulas of the previous section for the static limit, and then giving without proof the corresponding generalization to the doublet case. The justification for the use of the static limit is the smallness of the lowest order correction found in the previous section.

In the static limit we have (cf. (53))

$$\rho^2 - 1 = F_s = - \frac{D}{\omega - \Delta} \tag{63}$$

(We can drop the subscript s from now on, since in this section we are always dealing with the static limit.)

If we omit the G in the denominator of (46), and use (50) and (63),

we find

$$\Delta^{\ell} = \frac{B}{3}(\frac{1-\rho^2}{\rho^2})\int G^2(\varkappa)d^3\varkappa$$

$$= \frac{B}{3}(\frac{1-\rho^2}{\rho^2})(\frac{\pi}{2\mathcal{L}^2})^{3/2} \tag{64}$$

where again $G = \exp(-\mathcal{L}^2\varkappa^2)$. If instead we use a sharp cutoff in config-uration space at a distance r_0, we find

$$\Delta^{\ell} = \frac{4\pi^2 B}{3\varkappa_0^3}(\frac{1-\rho^2}{\rho^2}) \tag{64a}$$

In case of (47), we will have to rearrange the denominator some-what to get the same result which was obtained in references 5 and 7 with the aid of the Arnous-Bleuler transformation. This appears here to entail approximations, but it is really more accurate than direct evaluation of (47) would be. First, we omit all factors of G from (47), since they are not needed to ensure convergence. With the notation $C\varkappa = \nu$, we can write the denominator as

$$\nu(\nu_\lambda - \nu + i\xi)\left\{\nu(\nu_\lambda - \nu + i\xi) + \frac{\nu_0^2}{2}(\rho^2 - 1)\right\} .$$

Next we replace ν_λ by ν_0; it is further assumed that the main contribution to the integral is from the region $\nu \approx \nu_0$, so that ν can be replaced by $1/2(\nu + \nu_0)$. The denominator now becomes

$$\frac{1}{4}[\nu^2 - (\nu_0 + i\xi)^2][\nu^2 - \rho^2\nu_0^2] .$$

With all these rearrangements, the integral is easily evaluated:

$$\Delta^t = -i\sigma (\rho - 1) \tag{65}$$

Hence, by combining (45), (64a), and (65), we obtain

$$\Delta = i\, \rho\, \sigma + \frac{4\pi^2 B}{3\, \hbar_o^{\,3}} \left(\frac{1 - \rho^2}{\rho^2} \right)$$

Equations (63), (66) can be solved for ρ for any value of w .

In the doublet case the procedure is similar, though it is complicated by the presence of the spin-orbit splitting. One may use either a simple generalization of the methods of references 4 and 5, or of reference 7, or of the earlier sections of this article. (Actually, we used the methods of references 4 and 5). We denote the frequencies of the ${}^2P_{1/2}$ and ${}^2P_{3/2}$ transitions by ν_1 and ν_2, respectively. Instead of (63), we find

$$\rho^2 - 1 = -\frac{D}{3} \left\{ \frac{1}{\nu_\lambda - \nu_1 - \Delta} + \frac{2}{\nu_\lambda - \nu_2 - \Delta} \right\} \tag{67}$$

The equation for Δ remains formally unchanged: (66) still holds, with B given by (50). μ is now understood as the orbital dipole matrix element in which spin effects are neglected. We have neglected $\nu_1 - \nu_2$ against ν_1, so the natural linewidth is treated as the same for both lines.

We have calculated the absorption curves for a number of points in the region of Tomiser's experiments, using eqs. (66) and (67) with $r_0 = 8\ \overset{\text{o}}{\text{A}}$, a value which is reasonable and seems to give good agreement.

In carrying out these calculations, it is convenient to define

$$\nu_o = 1/2\,(\nu_1 + \nu_2)\ ;$$
$$\delta = \frac{\nu_2 - \nu_1}{2\,\sigma}\ ;$$
$$\Gamma = \frac{6\,\pi\,c^3}{\nu_o^{\,3}}\ \frac{N}{V}\ .$$

Figure 2 shows a typical absorption curve, corresponding to $\Gamma = 109.78$ (T = 688.1°K, P =)0.503 mm Hg.) The comparison between

experimental and theoretical linewidths is plotted in figure 3. It will be observed that there is qualitative agreement for both components of the doublet, even though the theoretical widths are approximately proportional to the square root of the density (cf eq. (56)), while the experimental dependence appears to be linear. The theoretical widths obtained by the methods of reference 1 would be too small to be seen on a graph of the scale of fig. 3, so we are justified in claiming to have improved the agreement. In the following article, it will be shown that the discrepancy which remains may well be due to the failure of the approximation (16).

APPENDIX

We can introduce a cutoff into the ordinary electrostatic interaction between two charge densities ρ_1 and ρ_2 by writing

$$\Phi = \int \rho_1(\underset{\sim}{r}_1)\, \rho_2(\underset{\sim}{r}_2)\, \varphi(r_{12}) d^3r_1 d^3r_2$$

$$= \frac{4\pi}{V} \sum_{\underset{\sim}{\kappa}} \frac{G(\kappa)}{\kappa^2} \int \rho_1(\underset{\sim}{r}_1)\, \rho_2(\underset{\sim}{r}_2) \exp[i\,\underset{\sim}{\kappa}\cdot(\underset{\sim}{r}_1 - \underset{\sim}{r}_2)]\, d^3r_1 d^3r_2$$

$$= \frac{4\pi}{V} \sum_{\underset{\sim}{\kappa}} \frac{G(\kappa)}{\kappa^2} \left\{ \int \rho_1(\underset{\sim}{r}_1) \exp(i\,\underset{\sim}{\kappa}\cdot r_1) d^3r_1 \right\} \left\{ \int \rho_2(\underset{\sim}{r}_2) \exp(-i\,\underset{\sim}{\kappa}\cdot\underset{\sim}{r}_2) d^3r_2 \right\}$$

$$(A - 1)$$

Here the sum goes formally over all $\underset{\sim}{\kappa}$ satisfying periodic boundary conditions in the volume V. If $G = 1$, we have the usual electrostatic interaction. If, both charge distributions can be treated as dipoles, we make the replacement

$$\exp(i\,\underset{\sim}{\kappa}\cdot\underset{\sim}{r}) \cong 1 + i\,\underset{\sim}{\kappa}\cdot\underset{\sim}{r}$$

and find

$$\Phi = \frac{4\pi}{V} \sum_{\underset{\sim}{\kappa}} \frac{G(\kappa)}{\kappa^2}\, (\underset{\sim}{\kappa}\cdot\underset{\sim}{\mu}_1)\,(\underset{\sim}{\kappa}\cdot\underset{\sim}{\mu}_2). \tag{A-2}$$

The matrix elements corresponding to (A-2) are given by eq. (5). Now, we can determine an appropriate choice of G just as well by looking at its effect on (A-1) as by examining (A-2). In other words, we consider the effect of G on the function

$$\varphi(\underset{M}{r}) = \frac{4\pi}{v} \sum_{\underset{M}{\kappa}} \cdot \frac{G(\kappa)}{\kappa^2} \exp(i \underset{M}{\kappa} \cdot \underset{M}{r}). \qquad (A-3)$$

The criterion we use for relating G to a cutoff radius r_0 is

$$\lim_{r \to \infty} r \; \varphi(\underset{M}{r}) = 1, \qquad (A-4)$$

and

$$\varphi(0) = 1/r_0 . \qquad (A-5)$$

Thus $\varphi_{(r)}$ is to become constant as r becomes $\ll r_0$, causing Φ in (A-2) to become small; it is to behave as an ordinary electrostatic potential for $r \gg r_0$. If one inserts $G = \exp(-\alpha^2 \kappa^2)$ into (A-3), one finds (replacing summation by integration, of course)

$$\varphi(r) \approx 1/r \qquad (\kappa \gg \alpha),$$
$$\varphi(0) = \frac{1}{\alpha \pi^{1/2}} , \qquad (A-6)$$

From (A-5) and (A-6), we find

$$r_0 = \alpha \pi^{1/2} , \qquad (A-7)$$

which is the relation between the wave number and configuration space cutoffs mentioned in sec. V. This prescription will also be used in the following article. It is obviously not unique, however. For example, if we required consistency between (64) and (64a), we would get a different result.
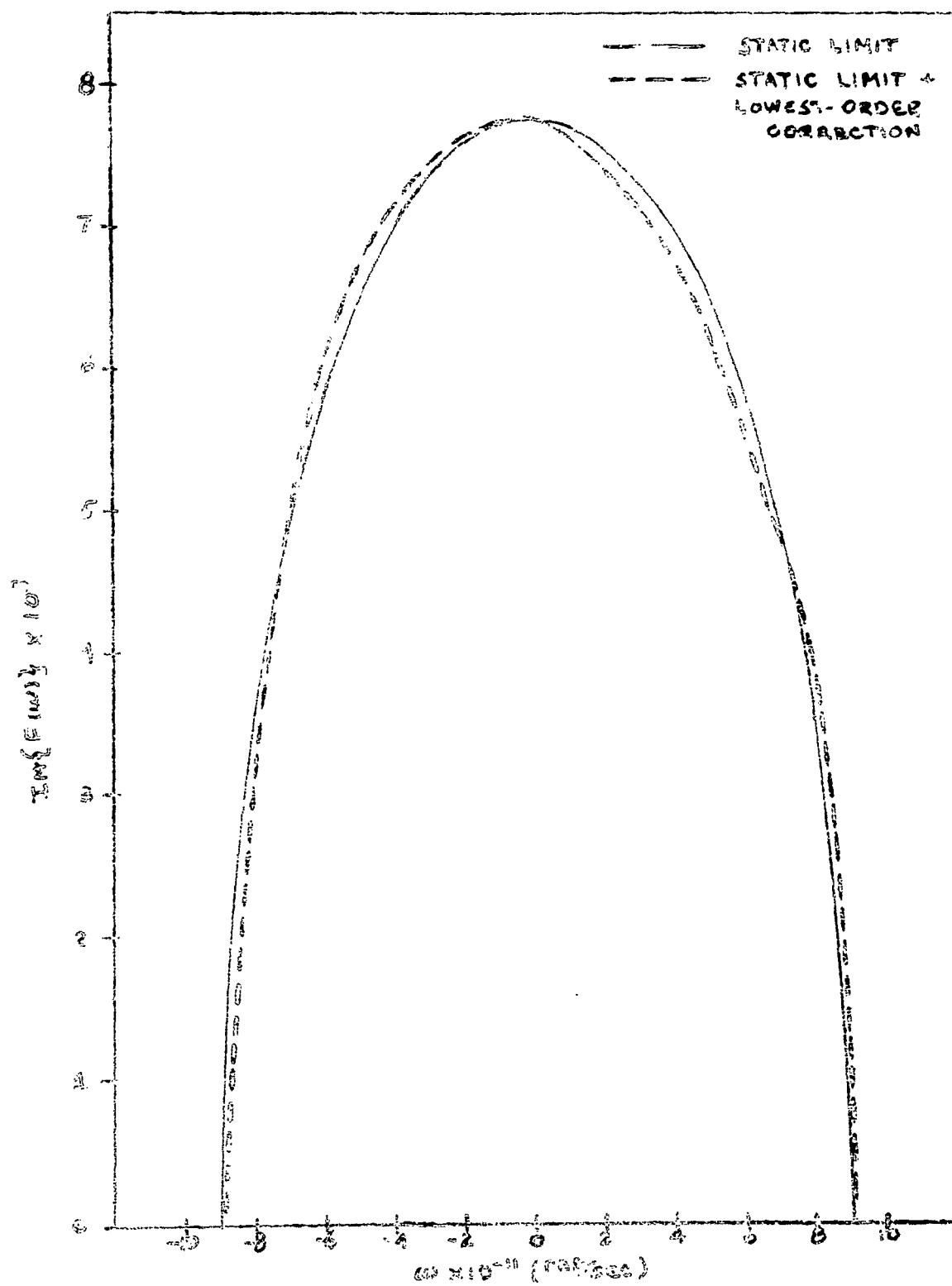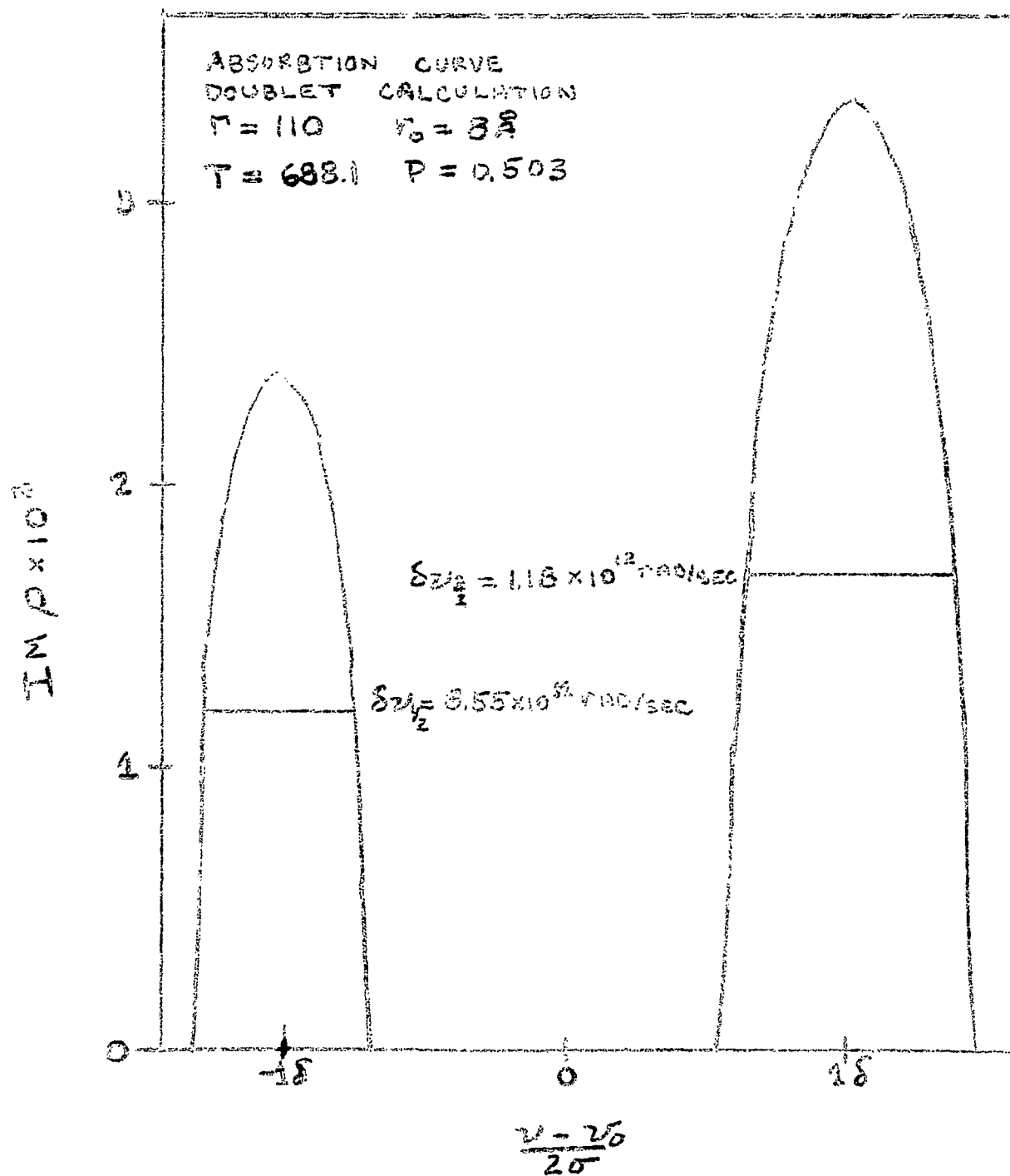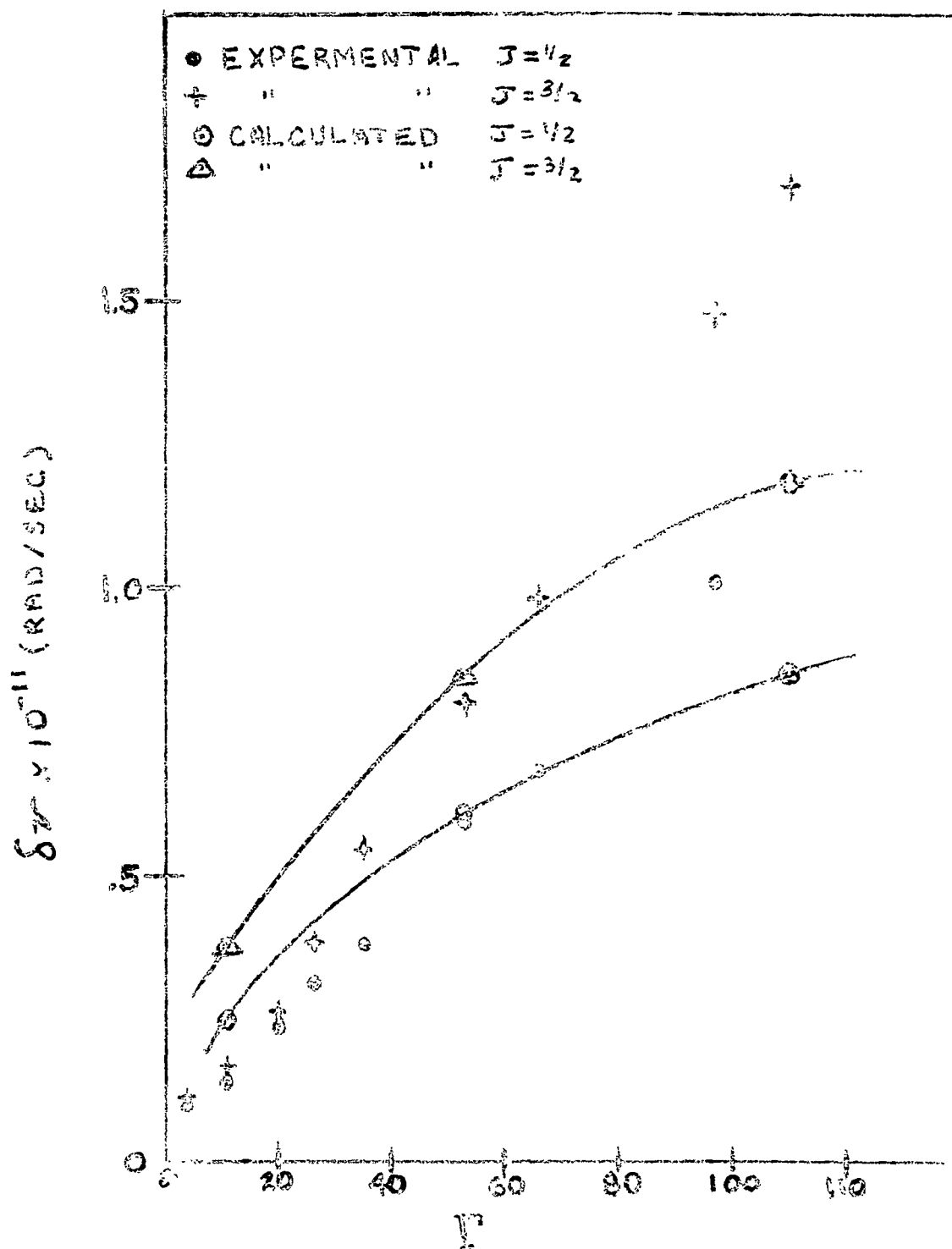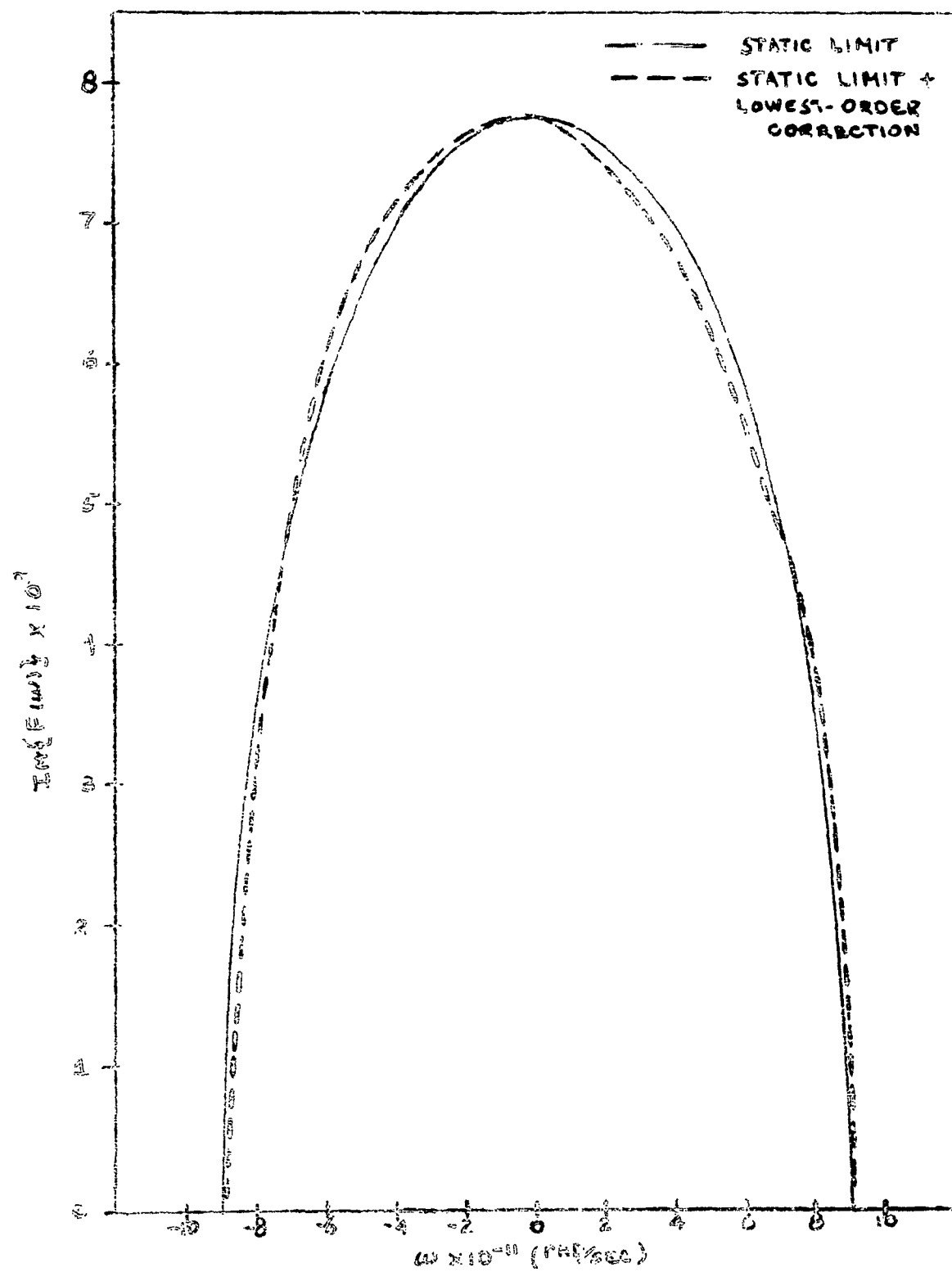
FIGURE 1

FIGURE 2

FIGURE 3

FIGURE 1

ABSORBTION CURVE
DOUBLET CALCULATION
$\Gamma = 110 \qquad r_o = 8 \overset{o}{A}$
$T = 688.1 \qquad P = 0.503$

$\delta \nu_{\frac{1}{2}} = 1.18 \times 10^{12} \, rad/sec$

$\delta \nu_{\frac{1}{2}} = 0.55 \times 10^{12} \, rad/sec$

$IM \, \rho \times 10^3$

$\dfrac{\nu - \nu_o}{2\sigma}$

FIGURE 2